Crystal Growth and Fluid Mechanics Problems in directional solidification

Final Progress Report

Saleh A. Tanveer, Gregory R. Baker, Michael R. Foster
Principal Investigators

July 1, 1996-June 30, 2000

The Ohio State University
Columbus, OH

Introduction
Our work in directional solidification has been in the following arenas:

1. Dynamics of dendrites including rigorous mathematical analysis of the resulting equations.

2. Examination of the near-structurally unstable features of the mathematically related Hele-Shaw dynamics

3. Numerical studies of steady temperature distribution in a vertical Bridgman device.

4. Numerical study of transient effects in a vertical Bridgman device.

5. Asymptotic treatment of quasi-steady operation of a vertical Bridgman furnace for large Rayleigh numbers and small Biot number in 3-D.

6. Understanding of Mullins-Sererka transition in a Bridgman device when fluid dynamics is accounted for.

1. Dendrite Dynamics

Over the period of support, we have developed and refined a new technique to study some features of the nonlinear evolution of a dendrite in the limit of small surface energy. The technique consists of extending the domain of the evolution equations to part of the complex plane in order to study the perturbatively small but singular surface energy effects. So, far we have restricted to a one-sided model for 2-D crystal growth that excludes complications of fluid motion or kinetic undercooling.

More specifically, we determined the evolution of singularities of the conformal mapping function $z(\zeta, t)$ that maps the upper-half plane into the exterior of a dendrite. For analytic initial conditions with isolated singularities, we related approach of complex singularities towards the real ζ-axis to observed interfacial features. Our concrete results include (i) predictions of certain classes of initial conditions for which arbitarily small surface-energy effects cause $O(1)$ perturbation in an other-wise smooth shape. (ii) prediction of time at such singular effects will be observed, (iii) prediction of interfacial scale dependence with surface energy for indentations associated with complex singularities, (iv) overall coarsening rate of side-branches over an intermediate distance from the tip that differs from the large distance asymptotics of Voorhees and Glicksman. These results based mostly on formal asymptotic and scaling arguments are presented in several long papers [1]-[3].

However, many important mathematical issues come up in the analysis. It is to be noted that the theory of complex higher order partial differential equations

2

in the complex plane is almost completely undeveloped– not much is known except the classic results of Cauchy-Kowaleski for first order equations. However, in the problems we encountered, the formal inner-outer asymptotic matching procedure sometimes requires us to have theoretical knowledge about existence of solutions of such parameter-free higher order complex partial differential equation. Formal scaling arguments that give rise to important physical results on interface dynamics, assume the existence of such solutions. We have now made serious progress to extend the general theory of complex partial differential equation in a fundamental way [4] that justifies and extends the formal results obtained previously in [1]-[3]. Earlier, we [5] had rigorously obtained results on singularities of similarity solutions. Together with existence and uniqueness results in [4], we now have a generic information on the location of complex singularities for a wide-variety of situations.

2. Examination of the near-structurally unstable aspects

Usually, in a physical problem, small terms of the equation are assumed to have small effects on the solution. When this fails, the problem is deemed to be structurally unstable. A structurally unstable system is of course unphysical, since a physically realistic system always involve small effects that are neglected in the model. What is not so clear a priori is what might be expected for a system that is not structurally unstable, but close to it. It has been argued in a recent review by one of us [6] that both viscous fingering for small surface tension, as well as dendrites for small anisotropy and small surface energy are examples of near structurally unstable system. For such systems, we have shown small terms in the equation that would seem reasonable to ignore can sometimes play a disproportionately large influence on the solution. Many surprising results in viscous fingering and dendrites have been interpreted in this way. In particular it is argued that convection and many other effects that are estimated to be small in any carefully designed dendrite experiment can be unexpectedly important, despite the estimates.

3. Numerical studies of steady Bridgman problem

We have continued further extending work [7] in this direction. Attention has been placed on resolving the influence of thin ampoule walls on the steady growth of crystals in a vertical Bridgman device. An asymptotic theory has been developed and tested against direct numerical simulation. There are three cases to consider.

We assume that in the regions of the ovens, a Newton law of heat transfer applies at the outer wall of the ampoule. We then verify that this law can be applied to the inner wall when the heat conductivities of the ampoule and the melt or crystal are of comparable magnitude. In other words the ampoule is transparent to the heat flow conditions.

For the region of the ampoule near the interface between a heater zone and the insulated region, there is an abrupt chnage in the heat transfer from a Newton law of heat transfer to no conduction. The thin ampoule has the effect of smearing the heat transfer coefficient so that a modified Newton law of cooling applies at the inner wall. An approximate version of this law was used

in our previous studies of steady crystal growth. The implication of the results is that thick ampoule walls lead to a degradation in radial segregation.

The third and final case has proved more problematic. This case occurs where the crystal interface meets the ampoule wall. Previous numerical studies have indictated that heat conduction in the ampoule causes the interface to curve near the wall. What is overlooked is the singular nature of the interface at the wall. We have found a similarity solution, but it does not meet the matching requirements for the temperature away from interface-wall region. Currently, a numerical study is being employed to identify the nature of the singularity. While finite element methods are often capable of treating singularities in the solution domain, it is possible that there are serious errors in previous numerical treatments by not taking the singular nature of the interface's contact with the ampoule in an appropriate way.

Our plan is to resolve the singular nature of the interface before completing three-dimensional studies.

4. Unsteady development of vertical Bridgman device

A highly efficient code has been used to study the transient behavior of the temperature distribution in the Bridgman device [5]. In particular, we have identified the requirements on the length of the ampoule in order to guarantee a phase of steady crystal growth. Direct comparison with popular one-dimensional models reveal their short-comings.

Some improvements have been made to the code: an explict Adams-Bashforth method is now applied to both the convective terms and the interface motion, while Crank-Nicolson is still used to treat the rapid diffusion of heat. Currently, tests are being conducted to to see whether direct solvers can replace the ADI splitting currently used in the code. If successful, and the prospects look good at the moment, direct solvers can then be applied for full three-dimensional simulations.

5. Asymptotic treatment of 3-D Bridgman with fluid motion

We have extended the previous results reported by Tanveer ['94, Phys. Fluids A] and Foster [8] for axi-symmetric situations into two ways. First, we have made the analysis fully three-dimensional, allowing for ampoule about which there is an azimuthal variation in the heat transfer through the wide walls, or azimuthal variation due to a slight tilt of the ampoule axis [9] We find that the three-dimensional effects in every case increase the horizontal segragation in the crystal. We also determined that the optimization criteria found first by Tanveer extend in very similar form to the thee-dimensional situation, but now involve zeros of the J_1 Bessel function rather than J_0. Secondly, we have explored effects of rapid rotation of the ampoule about the vertical axis [10]. We find, in agreement with other recent numerical work, that in fact rotation of the ampoule *worsens* the radial segregation in the crystal. What effect that rotation has on the onset of morphological instability awaits further work.

6. Mullins-Sererka transition in Bridgman device

We have continued the effort on Mullins-Sererka instability in a Bridgman device in the large Ra limit; we find that there is a fundamental difference in the results from that obtained in an unrestricted geometry that several workers have found by neglecting convection altogether. Nonlinear results include the possibilities of finite-time singularity or algebraic growth in time in the nonlinear $Ra^{-1/6}$ boundary layer at the interface between the melt and the crystal. Final publication of this work awaits further confirmation of the numerical results obtained by a spectral scheme.

References

[1] M.D. Kunka, M.R. Foster & S. Tanveer, "Dendritic Crystal Growth for weak undercooling", *Physical Review E*, 56, pp 3068-3100, 1997.

[2] M.D. Kunka, M.R. Foster & S. Tanveer, "Dendritic Crystal Growth for weak undercooling. Part II: Surface energy effects on nonlinear evolution", *Physical Review E*, 59, pp 673-710, '99.

[3] M.D. Kunka, M.I. Siegel & S. Tanveer, "Singular surface energy effects for anisotropic surface energy for a of a neede crystal and a Hele-Shaw interface", under preparation, '00.

[4] A. S. Fokas & S. Tanveer, "A Hele-Shaw Problem and the Second Painleve' Transcendent", *Math. Proc. Camb. Phil. Soc.* 124, pp 169-191, 1998.

[5] O. Costin & S. Tanveer, "Existence and Uniqueness for a class of nonlinear higher-order partial differential equations in the complex plane", Submitted to Communications in Pure and Applied Math, 1999.

[6] S. Tanveer, "Surprises in viscous fingering", To appear in the *Journal of Fluid Mechanics*

[7] A numerical study of steady crystal growth in a vertical Bridgman device, Miklos Jalics, Ph.D dissertation, The Ohio State University, 1998.

[8] M. R. Foster, "Asymptotic analysis of a vertical Bridgman furnace at large Rayleigh number", *Physics of Fluids* 9, pp 683, 1997.

[9] M.R. Foster, 1999 "Asymptotic Analysis of a Three-Dimensional Bridgman Furnace at Large Rayleigh Number", *Phys. Fluids*, 11, pp 1827.

[10] M. R. Foster, 2000 "The effect of rotation on a vertical Bridgman growth at large Rayleigh number", *J. Fluid Mech.*, 409, pp 185

THE OHIO STATE UNIVERSITY

March 16, 2001

Ms. Deborah T. King
NASA Glenn Research Center
Space Systems & Grants Branch
Mail Stop 500-319
21000 Brookpark Road
Cleveland, OH 44135-3191

Re: **Final Technical Report for Grant No. NAG3-1947; The Ohio State University**
       Research Foundation No. 732118

Dear Colleague:

On behalf of the Principal Investigator, Dr. Saleh Tanveer of the Department of Mathematics,
The Ohio State University Research Foundation submits the Final Technical Report for Grant No.
NAG3-1947.

Should you require additional administrative information please contact Dave Davis, the
Sponsored Program Officer for this project, by phone 614.292.4326, fax 614.292.4315, or email
davis.60@osu.edu. Questions regarding the technical aspects of the project should be directed to
Dr. Tanveer by phone 614.292.5710, or email tanveer@math.ohio-state.edu. Thank you for your
support of this research effort.

Sincerely,

Amy B. Dudley
Sponsored Programs Associate
Office of Grants and Contracts

cc: CASI
       Arnon Chait
       Office of Naval Research

Enclosure